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APPLICATION NUMBER: ***60/514,949***
FILING DATE: ***October 28, 2003***
RELATED PCT APPLICATION NUMBER: ***PCT/US04/35573***

Certified by

Jon W Dudas

Acting Under Secretary of Commerce
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PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

Express Mail Label No. ET905047335US

INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Neal	Kalechofsky	Stow, Massachusetts

☐ Additional inventors are being named on the _______ separately numbered sheets attached hereto

60/514949 102803

TITLE OF THE INVENTION (500 characters max)

A DEVICE TO SORT, SEPARATE, AND SIZE PARTICLES

Direct all correspondence to: **CORRESPONDENCE ADDRESS**

☐ Customer Number [ ] *Type Customer Number here* → *Place Customer Number Bar Code Label here*

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ENCLOSED APPLICATION PARTS ***(check all that apply)***

☑ Specification *Number of Pages* 12
☐ Drawing(s) *Number of Sheets*
☐ Application Data Sheet. See 37 CFR 1.76
☐ CD(s), Number
☐ Other (specify)

METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☐ Applicant claims small entity status. See 37 CFR 1.27.
☑ A check or money order is enclosed to cover the filing fees
☐ The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number:
☐ Payment by credit card. Form PTO-2038 is attached.

FILING FEE AMOUNT ($): $160.00

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.
☑ No.
☐ Yes, the name of the U.S. Government agency and the Government contract number are: ____________

Respectfully submitted,

SIGNATURE [signature]

Date 10/28/2003

TYPED or PRINTED NAME Todd S. Parkhurst

REGISTRATION NO. *(if appropriate)* 26,494

TELEPHONE 312-578-6694

Docket Number: Oxford Instrumen

USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

10/28/03 15541 U.S. PTO

FEE TRANSMITTAL for FY 2004

Effective 10/01/2003. Patent fees are subject to annual revision.

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT ($) 160.00

Complete if Known	
Application Number	
Filing Date	
First Named Inventor	Neal Kalechofsky
Examiner Name	
Art Unit	
Attorney Docket No.	

METHOD OF PAYMENT *(check all that apply)*

☑ Check ☐ Credit card ☐ Money Order ☐ Other ☐ None

☑ Deposit Account:

Deposit Account Number: 50-1794

Deposit Account Name: Holland & Knight LLP

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FEE CALCULATION

1. BASIC FILING FEE

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1001	770	2001	385	Utility filing fee	
1002	340	2002	170	Design filing fee	
1003	530	2003	265	Plant filing fee	
1004	770	2004	385	Reissue filing fee	
1005	160	2005	80	Provisional filing fee	160.00

SUBTOTAL (1) ($) 160.00

2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE

			Extra Claims		Fee from below		Fee Paid
Total Claims		-20** =		X		=	
Independent Claims		- 3** =		X		=	
Multiple Dependent						=	

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description
1202	18	2202	9	Claims in excess of 20
1201	86	2201	43	Independent claims in excess of 3
1203	290	2203	145	Multiple dependent claim, if not paid
1204	86	2204	43	** Reissue independent claims over original patent
1205	18	2205	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) ($) 0.00

**or number previously paid, if greater; For Reissues, see above

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1051	130	2051	65	Surcharge - late filing fee or oath	
1052	50	2052	25	Surcharge - late provisional filing fee or cover sheet	
1053	130	1053	130	Non-English specification	
1812	2,520	1812	2,520	For filing a request for *ex parte* reexamination	
1804	920*	1804	920*	Requesting publication of SIR prior to Examiner action	
1805	1,840*	1805	1,840*	Requesting publication of SIR after Examiner action	
1251	110	2251	55	Extension for reply within first month	
1252	420	2252	210	Extension for reply within second month	
1253	950	2253	475	Extension for reply within third month	
1254	1,480	2254	740	Extension for reply within fourth month	
1255	2,010	2255	1,005	Extension for reply within fifth month	
1401	330	2401	165	Notice of Appeal	
1402	330	2402	165	Filing a brief in support of an appeal	
1403	290	2403	145	Request for oral hearing	
1451	1,510	1451	1,510	Petition to institute a public use proceeding	
1452	110	2452	55	Petition to revive - unavoidable	
1453	1,330	2453	665	Petition to revive - unintentional	
1501	1,330	2501	665	Utility issue fee (or reissue)	
1502	480	2502	240	Design issue fee	
1503	640	2503	320	Plant issue fee	
1460	130	1460	130	Petitions to the Commissioner	
1807	50	1807	50	Processing fee under 37 CFR 1.17(q)	
1806	180	1806	180	Submission of Information Disclosure Stmt	
8021	40	8021	40	Recording each patent assignment per property (times number of properties)	
1809	770	2809	385	Filing a submission after final rejection (37 CFR 1.129(a))	
1810	770	2810	385	For each additional invention to be examined (37 CFR 1.129(b))	
1801	770	2801	385	Request for Continued Examination (RCE)	
1802	900	1802	900	Request for expedited examination of a design application	

Other fee (specify) ______________

*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) ($)

SUBMITTED BY				(Complete *(if applicable)*)	
Name (Print/Type)	Todd S. Parkhurst	Registration No. (Attorney/Agent)	26,494	Telephone	312-578-6694
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Title:

A Device to Sort, Separate, and Size Particles

Abstract:

A novel technique that uses the unique properties of superfluid helium as a viscous medium in which to sort, separate, or size particles (typically, but not necessarily, of very small average dimension) is described. The principal application for the technique is to sort/size particles with average diameters small enough (approximately less than 3 microns) that existing techniques are ineffective.

Motivation:

Mechanisms to size (determine their average diameter in a sample) and/or classify particles (sort them as a function of size) are fundamental to powder processing technology and at the heart of a wide number of industrial applications. Examples include nanotechnology, pharmaceuticals, High Performance Liquid Chromatography (HPLC) column manufacture, abrasives, foodstuffs and many others.

Many of these industries are increasingly interested in techniques that can size/classify particles on an ever decreasing length scale. Conventional separation techniques, described in more detail below, tend to become ineffective when the average particle size in a dry powder or slurry (ie, particles in solution) is less than approximately 3 microns in diameter.

As an example of this, emerging applications in HPLC require that the LC column be filled with silica particles with average diameters on the order of 1 micron in size and with a size distribution less than 20%. Current classification techniques are unable to satisfy this requirement on an industrial scale and the industry is actively seeking to develop methods to achieve this.

The market for LC columns is approximately $100 million per year in the US alone. Next generation techniques are expected to be a growing segment of this market as the applications for micro and nano LC increase.

Existing Methods of Particle Sizing/Classification

For powders or slurries (powders in solution) whose average particle size is greater than ~ 3 microns well established methods to classify the individual particles exist. For example, commercially available air classifiers employ the well known property that particles falling through a gravitational field (either the Earth's, or the artificially enhanced gravitational field of a centrifuge) in a viscous medium will exhibit a size dependent terminal velocity. Specifically, particles will achieve a terminal velocity that goes as the square of their respective diameters and the inverse of the viscosity of the separation medium (Stokes settling):

$$V_T = (2/9)*(d^2)*g*(p_1 - p_0)/\mu$$

Where air at STP is used as the viscous medium, this method works reliably down to ~ 3 micron in average particle diameter (Fig 1). Once the particles are spatially well separated they may be readily sized or sorted.

For samples consisting of powders or slurries with smaller average particle diameters, however, traditional methods begin to encounter problems in achieving separation between particles. In increasing importance, these are:

1) The terminal velocity of the particles becomes very low, leading to unreasonably long separation times
2) Random scattering due to thermal fluctuations in the viscous medium dominate particle dynamics (Brownian diffusion). This tends to randomize particle motion and mix larger particles back in with smaller ones

3) Surface to volume ratios are so large that surface attractions (van der Waal forces) cause particles to agglomerate. For example, silica particles with diameters less than 2 microns will agglomerate in room temperature air, forming larger particles that cannot be easily separated and classified.

Depending somewhat on the properties of the particles to be classified, the limitations imposed by these realities tend to set in around the 3 micron range (see Fig 1). Not surprisingly, this is the point at which many powder processing applications have remained at a standstill, waiting for some method to permit particles with smaller diameters to be classified.

The Innovation:

Replace the viscous medium typically employed by air classifiers, air at 300 Kelvin temperatures, with superfluid helium at $T < 2.2$ Kelvin

Properties of Helium

Helium is an inert gas at STP. At 4.2 Kelvin, under its own vapor pressure, it liquifies. If liquid helium is further cooled to less than 2.2 K, it undergoes a further phase transformation to what is commonly known as the "superfluid" state (see Fig 2). The properties of superfluid helium are well documented in the literature and include:

1) A very high thermal conductivity
2) A very high specific heat
3) A very low, and strongly temperature dependent, viscosity
4) Zero boiling
5) A very high wetting coefficient

These properties can be exploited to make superfluid helium extremely attractive as the basis of a particle separation device. The intrinsically low temperature of superfluid helium limits the deleterious effects of Brownian diffusion. The low, and strongly temperature dependant, viscosity implies a rapid and controllable terminal velocity in settling particles. Finally, superfluid helium's high wetting coefficient, combined with its very high thermal conductivity, allows it to act as an effective low temperature "surfactant" and deter particle agglomeration.

The reason for this last, and important, point may need further clarification. Due to superfluid helium's high thermal conductivity/high specific heat, particles immersed in superfluid helium rapidly cool. Once particles are at ambient temperature, due to helium's high wetting coefficient, layers of helium adhere to the particle surface. Thus particle-particle van der Waal (VDW) attraction—the basis for most agglomeration events—is deterred because the particles are prevented from physically coming very close to one another. Instead, the intervening layers of helium keep particles apart and minimize VDW agglomeration (Fig. 3a and b). VDW attraction is a very strong function of interparticle distance, varying inversely d^6. Thus even a small increase in interparticle distance implies a drastically reduced agglomeration rate.

The fundamental claim, therefore, should protect the use of superfluid helium as a viscous medium to sort, separate, size or classify particles. A secondary claim should include ordinary liquid helium as well as the other cryogenic liquids/fluids or low temperature gas (note, however, that I doubt that these other liquids, fluids or gases could in fact be used in actual applications. Cryogenic fluids boil, which would distort particle motion, and in any case particles can be expected to agglomerate in any liquid EXCEPT helium. Nonetheless our claim should extend to them just to be safe)

An additional claim should protect the use of superfluid helium, ordinary liquid helium, or other cryogenic fluids/liquids as a surfactant in particle storage and/or low temperature chemistry applications.

Applications of the Innovation

Once the fundamental innovation—using superfluid helium as a separation medium in which to classify particles—has been accepted, a number of useful applications may be envisioned.

1) **Particle Sizer**—being a device designed to separate small amounts of particles such that the average diameter of the particles in a given sample may be determined
2) **Particle Classifier**—being a device designed to separate large amounts particles such that they may be sorted for use in a variety of applications.

In both cases, the envisioned process takes place in a number of distinct operations:

1) Powder samples, either dry or in a slurry, are baked out to remove air/solution
2) Particles are cooled to near liquid helium temperatures by being mixed into a helium gas stream and forced through a precool tube grounded to increasingly colder thermal baths. This also has the effect of separating previously agglomerated particles (particles with diameters less than 2 microns can be expected to agglomerate in air at room temperature) via collisions between the agglomerated particles and the walls of the precool tube
3) Particles are sprayed onto the surface of superfluid helium and separate as they settle at different velocities through the superfluid

Figures 4 – 6 show some preferred embodiments of machines designed to achieve one or another of the above applications. A step by step description of the process in each instance is given below

1) Powder samples, either dry or in a slurry, are placed in a seeder. The seeder should be vacuum sealable, insulated and bakeable to temperatures up to 150 degrees Celsius. It should be fitted with a heater and a pumping port to allow air and solution to be driven out of the seeder. Valve 1, 2, 3, 4 are closed
2) The seeder volume is heated using the heater. Valve 2, 4 are opened and air/solution in the seeder volume pumped away
3) The heater is turned off, Valve 4 is closed and valve 3 opened to the helium gas supply. A small pressure of helium gas is maintained while valve 1 is opened. Particles are driven from the seeder and into the precool tube.
4) The precool tube is caused to run through a series of heat exchangers where the particle/helium gas stream is cooled to increasingly lower temperatures. In a preferred embodiment, this can be done by the particle/gas stream through a series of heat exchangers consisting of many loops of precool tube (not shown for clarity in Fig 4). The heat exchangers are in turn thermally grounded to increasingly colder thermal baths. The thermal baths can be provided by stored cryogenic liquids or, in a preferred embodiment, by a cryogen free refrigeration device, such as a closed cycle cooler or a pulse tube cooler, capable of producing very low temperatures with little or no use of liquid helium.

   The cooling stages shown in Fig 4 - 6 consist of 77 K (the temperature of liquid nitrogen) and 5 K (just above the temperature at which helium liquifies). This temperature sequence is only intended as an example and is not exhaustive of the temperature staging that may be employed to precool the particle stream before entering the superfluid bath

5) As agglomerated particles travel through the loops of precool tube they suffer many collisions that should be violent enough to deagglomerate them.
6) As the deagglomerated particles exit the last heat exchanger they should be well cooled to the temperature of the exchanger. Thus their specific heat will be very small compared to that of helium at these temperatures. Hence when they exit the precool tube and impact on the superfluid helium in the separation tube there should be little boiling and temperature gradient imposed on the superfluid.

At this point the further operation/construction of the unit depends on whether the desired application is to size the particles or to classify them. Since there are useful industrial applications for both, I will describe two units that employ the basic innovation to achieve the desired end

Particle Sizer:

After being cooled to 5 K, the particles exit the precool tube directly over the surface of the superfluid in the separation tube. The separation tube is U shaped for reasons that shall be made clear below. The superfluid tube is kept at $T < 2.2$ K by contact with a thermal bath at this temperature. Preferably this can be produced via a refrigeration device such as a closed cycle cooler or a pulse tube, although other refrigeration arrangements can be envisioned as well. The tube is partially filled with superfluid helium with the region above the superfluid filled with low pressure helium gas.

The diameter of the separation tube is caused to be wider than the precool tube to reduce collisions between the particles and the walls of the separation tube.

Particles emerge from the precool tube in a stream of helium gas and impact on the surface of the superfluid. Due to superfluid helium's superior properties as a separation medium, as described above, they rapidly separate in the vertical direction. Once well separated, the particles may be sized using a number of well understood techniques. As a preferred embodiment, a laser is used to produce a diffraction pattern on a screen. The diffraction pattern can be devolved to yield size information on the particles in the beam falling through the superfluid.

Since sample sizes in a typical sizing experiment are very small, it might be acceptable to simply leave the particles to accumulate in the bottom of the superfluid tube. Occasionally the tube would need to be heated to room temperature and cleaned to remove accumulated particles.

As a preferred alternative, and as a potentially separate innovation, one could employ a flush method to cleanse the unit of particles on a regular basis. Once the laser indicates that no more particles are falling through the superfluid, particles may be flushed from the system as follows. A pressure of helium gas over the supefluid helium forces the liquid helium and particles in solution in the separation tube out through the exit U shaped tube. At a predetermined height above the original liquid helium level a flush stick, consisting of a stainless steel stick with a high surface area sinter on the end of it, is placed. The liquid helium is forced through the sinter by the gas pressure, and all of the particles in the liquid are plated out on the surfaces of the sinter. The pressure is then released, causing the liquid to flow back through sinter at the end of the flush stick once more (thus depositing any particles that may have escaped during the initial flush). The liquid returns to its original level below the flush stick. The stick can then be withdrawn up to room temperature as desired, and the particles adhered to the sinter's surfaces driven off and recaptured.

The flush stick can also be held at a slight voltage to further attract particles.

Particle Classifier

In this approach the goal is to physically separate the particles, rather than simply size them. Two preferred embodiments, based on the basic innovation of using superfluid helium as a viscous medium, can be described

A: As described above, the diameter of the tube is widened to allow the particles to fall through the superfluid bath without suffering further collisions with the walls of the bath. The entry port of the precool tube is placed well to one side of the superfluid bath, entering the bath in the vertical direction.

A magnetic gradient field is maintained across the superfluid bath in the lateral direction. The action of the magnetic gradient causes larger particles, which experience a higher degree of magnetic repulsion (note the assumption here is that the particles are diamagnetic. In the case of para or ferro magnetic particles, an

inverse operation may be used). Larger particles are pushed to the far side of the chamber, while smaller ones tend to fall nearer to the position of the entry of the precool tube.

The bottom of the chamber is divided into bins. As described above, each bin may be fitted with an individual exit tube/ flush stick arrangement such that particles may be separately captured and returned to room temperature.

B In an alternate embodiment, the entry port of the precool tube is placed well to one side of the superfluid bath, entering the bath in the lateral direction. Particles emerge from the precool tube with, initially, only lateral velocity. The action of gravity in the vertical direction causes particles to fall into the superfluid and, once immersed, acquire a size dependent terminal velocity in the vertical direction. Those with higher terminal velocities in the vertical direction will tend to fall first, and hence separation in the lateral direction can be readily achieved.

As before, the bottom of the chamber is divided into bins. As described above, each bin may be fitted with an individual exit tube/ flush stick arrangement (for clarity only 2 are shown in the accompanying figure) such that particles may be separately captured and returned to room temperature.

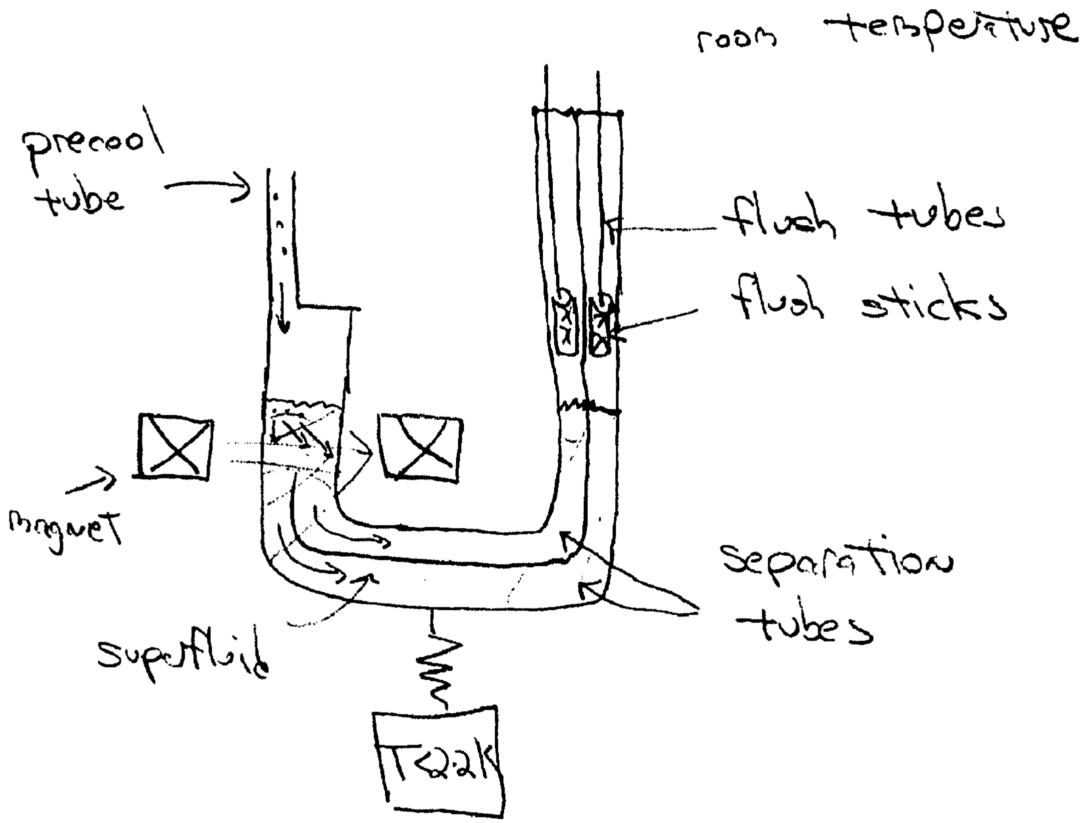


Figure 6: Sketch of a preferred embodiment of a particle classifier based on the proposed innovation. See text for details.

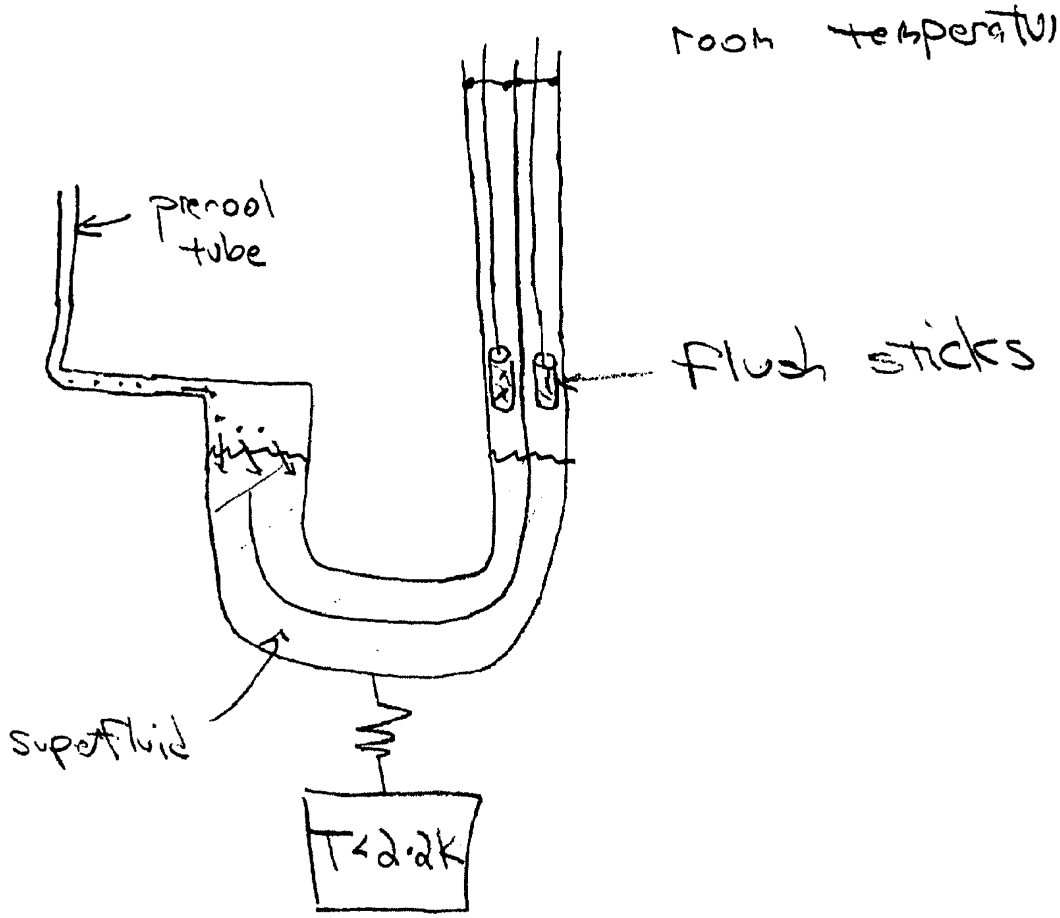


Figure 7: Sketch of an alternate preferred embodiment of a particle classifier based on the proposed innovation. See text for details.

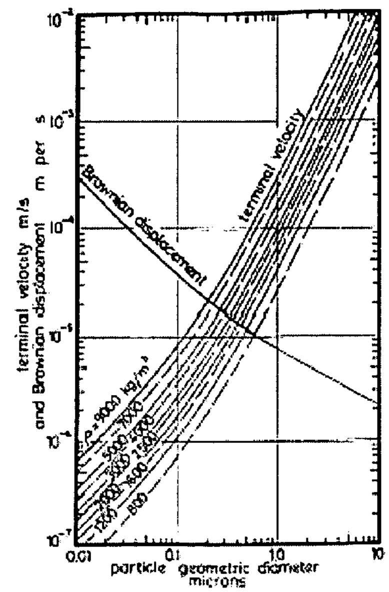
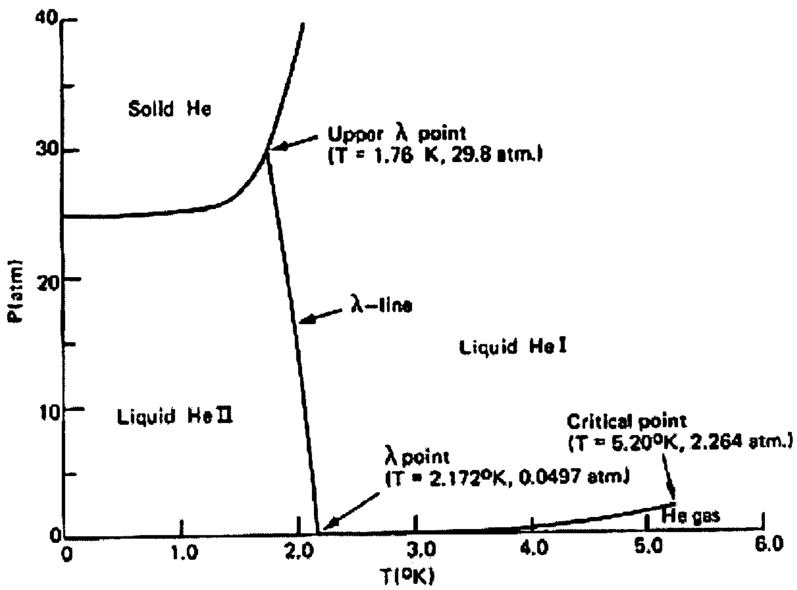


Figure 1: Terminal velocities and Brownian displacements of various particles falling through still air with $g = 9.8\ m/s^2$



The phase diagram of He^4.

Figure 2: Phase diagram of 4He. Note phase transformation to HeII superfluid state (so called "lambda point") at T = 2.2 Kelvin at saturated vapour pressure

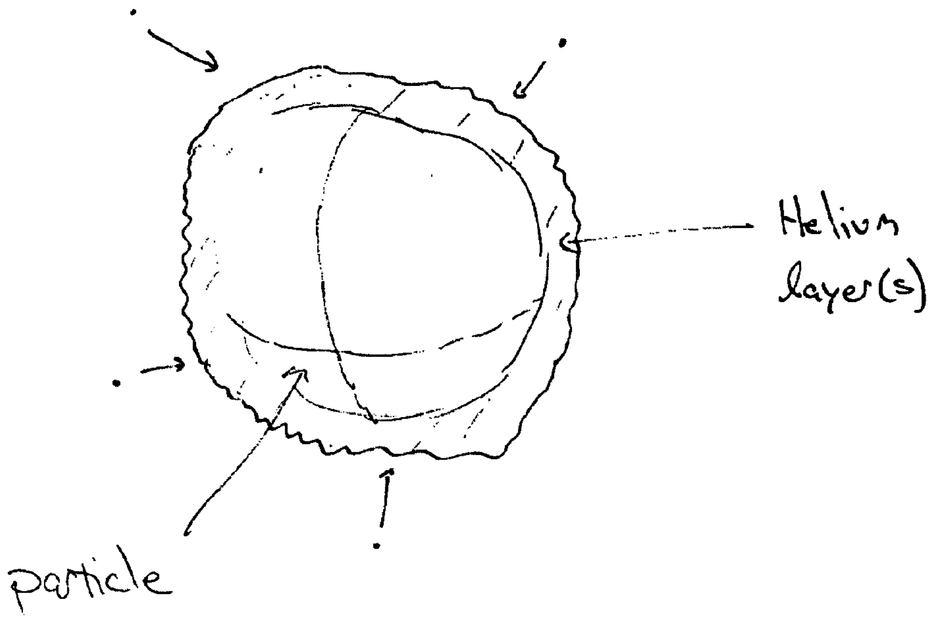


Figure 3a: A cold particle in superrfluid 4He. 4He atoms are drawn from the aurrounding fluid and, being more attracted to the particle than to themselves, adhere and form layer(s) on the particles surface

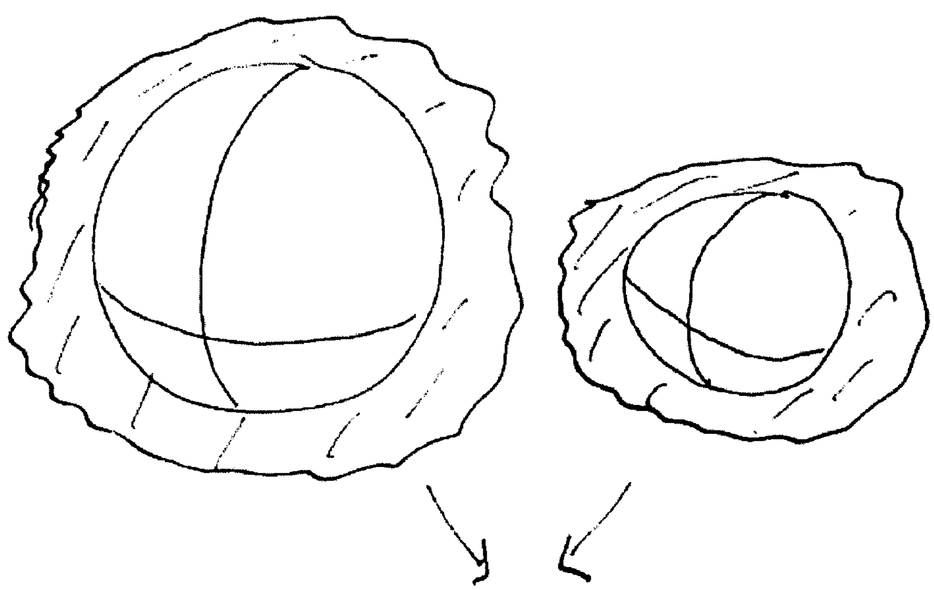


Figure 3b: Two particles, insulated from one another by layer(s) of adhered helium atoms, scatter off one another in the background of the superfluid. The VDW between the particles (and the helium layers) is very weak and agglomeration between the particles is deterred.

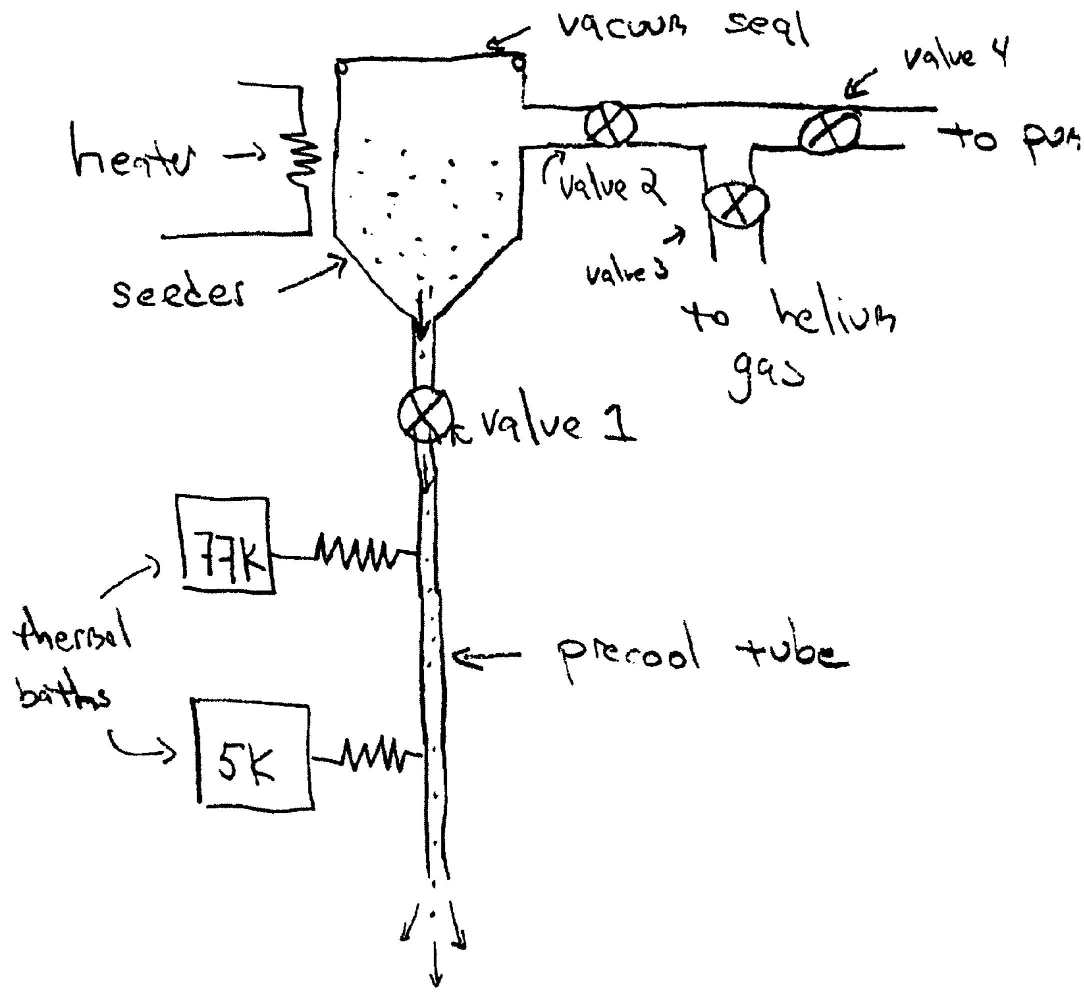


Figure 4: The seeder and precool stage of a device that may be used as a first stage for a superfluid helium based particle sizer and/or classifier. Note loops in precool tube are not shown for clarity

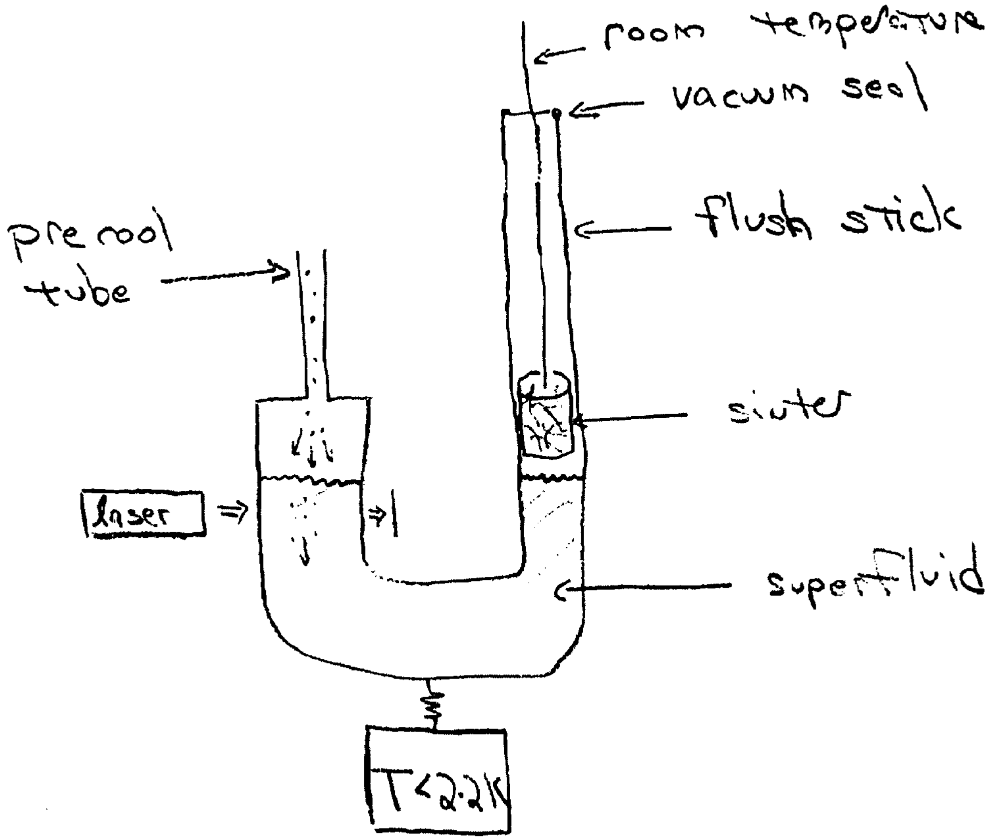


Figure 5: Sketch of a particle sizing device based on the proposed innovation. See text for details